

A Beamline to National Security

Researchers at the Center for Accelerator Mass Spectrometry continually find new ways to advance Laboratory science.

Livermore researcher Scott Tumey adjusts the controls for a nuclear forensics experiment at the Center for Accelerator Mass Spectrometry (CAMS).

THE Center for Accelerator Mass Spectrometry (CAMS) at Livermore is world renowned for highly accurate dating of trees, ice, corals, archaeological samples, and human tissues. CAMS grew out of the construction in the mid-1980s of a multipurpose tandem accelerator laboratory designed for applications that use ion-beam analytic techniques. Over the years, accelerator mass spectrometry (AMS) became the largest component of the center's work, and today, CAMS is the most heavily used AMS facility in the world.

AMS replaces the older decay-counting methodology, which could take several days to complete, with a precise count of rare isotopes sometimes available within seconds. Although carbon-14 is the most commonly used isotope for dating samples, CAMS can also measure aluminum, beryllium, calcium, chlorine, hydrogen, iodine, plutonium, strontium, tritium, and uranium. The unique sensitivity of CAMS has led to groundbreaking research in earth and environmental sciences; climate change; and human metabolism of chemicals, toxic compounds, and nutrients.

Less well known is research on behalf of the Laboratory's primary mission, national security. "Although our national security work hasn't received a lot of play, the accelerator has been applied to forensics studies since the center was established in 1988," says Graham Bench, director of CAMS, which is part of Livermore's Physical and Life Sciences Directorate.

Forensics to Protect Human Health

In the late 1990s, CAMS scientists helped solve a long-standing controversy about the neutron dose received by survivors of the atomic bomb dropped on Hiroshima in 1945. Health data from survivors of the Hiroshima and Nagasaki bombings are used to calculate the risks of radiation-induced cancers in humans.

However, low-energy measurement data at the time indicated that the neutron dose estimate published in a frequently cited 1986 report was too low.

Researchers at CAMS had by then developed a method for detecting trace amounts of radioactive nickel-63 in copper. They applied that technique to copper samples taken from locations ranging from 380 to more than 5,000 meters from the explosion's center, including the Bank of Japan, a soy sauce brewery, an elementary school, and the roof of a Shinto shrine. The team's findings provided the first clear measurements of the neutron dose to survivors in Hiroshima, which essentially agreed with the 1986 estimate.

Anthrax attacks in Washington, DC, and Florida prompted the Federal Bureau of Investigation to call on CAMS in 2001. The bureau needed to determine the age of spores found on envelopes and other contaminated objects. Results from carbon-14 dating experiments indicated the spores had been produced within 12 months of the attacks, information that helped determine possible production sites for the anthrax.

In addition, for more than 10 years, CAMS measurements have helped monitor urine samples from cleanup workers on Rongelap Atoll in the Marshall Islands to ensure that plutonium exposure does not exceed the established safety standards. The Rongelap community had been displaced to a neighboring atoll in 1954 to protect residents from radioactive fallout following an atmospheric nuclear test over nearby Bikini Atoll. The U.S. government resettled the islanders back to Rongelap in 1957. However, in 1985, the community relocated again because of their concerns about lingering contamination. Today, workers at Rongelap are remediating the soil to reduce levels of radioactivity in the atoll's plants. (See *S&TR*, July/August 2010, pp. 18–21.)

Nuclear Forensics Benefits

The nonproliferation community gained a new appreciation for CAMS during a 2009 nuclear forensics exercise. When beryllium-10 testing was requested as part of the exercise, the center was called in to help. CAMS had been using beryllium-10 as a dating tool for the geosciences since the 1990s but had not previously applied the technique for nuclear forensics.

Livermore researcher Scott Tumey leads the center's nuclear forensics and heavy isotope research. He and Tom Brown pioneered many developments in accelerator mass spectrometry and over the years have optimized new uses for the spectrometers.

For example, CAMS was one of the first facilities with the capability to measure strontium-90, which is present in nuclear test fallout. Since then, Tumey and Brown have enhanced the strontium detector, increasing its sensitivity by a factor of almost 100. The improved sensitivity allows researchers to more accurately assess the effectiveness of remediation work in the Marshall Islands. Similarly, for uranium-233, Tumey's modifications to a previous measurement setup increased sensitivity by a factor of 1,000 for more accurate nuclear forensics.

According to Bench, the center now has more projects in support of national security than ever before. "We're developing techniques that will open new frontiers of nuclear science for the Laboratory," he says. "We're also finding more applications for many of our isotope measurements. For example, we are beginning to apply our aluminum-26 capabilities, which we've used for years in geologic studies, to nuclear forensics."

New Beamline at Work

A CAMS project completed this year examined the helium bubbles that form in weapons-grade plutonium as it ages.



Laboratory scientists (from left) William (Skip) Fields, Karis McFarlane, and Scott Tumey prepare for an experiment on the high-energy ion-implantation beamline at CAMS. The new beamline is designed to irradiate highly radioactive elements used in nuclear weapons and to fuel nuclear power plants.

As a nuclear weapon sits in the stockpile, plutonium decays into a uranium atom with the emission of a helium nucleus. The helium nuclei create vacancies in plutonium's solid structure and may coalesce to form voids that could weaken the material. To better examine plutonium aging, researchers used a new high-energy ion beamline to implant helium ions in minute samples and mimic how helium builds up during the natural decay process.

Ion beamlines have been in place at CAMS since the late 1980s when they were used for proton tomography and to initiate nuclear reactions in materials for radiation detector testing and calibration. Ion implantation is a common practice in semiconductor manufacturing, but these industrial applications use relatively low energies such that ions penetrate less than 1 micrometer. The CAMS accelerator can achieve energies up to

tens of megaelectronvolts to implant ions hundreds of micrometers deep. The new ion beamline is designed to irradiate actinides, a group of highly radioactive elements, many of which are used in nuclear weapons and to fuel nuclear power plants.

"The helium-ion implantation project ran for five years," Bench says, "and required both the ion beamline and radiation shielding for the target chamber." The shielding, designed to attenuate radiation, is constructed of 5 centimeters of lead surrounded by 70 centimeters of polyethylene. It rides on rails for placement over the target chamber during an experiment. With this heavy shield reducing radiation exposure, higher ion-beam currents can be aimed at a plutonium sample.

Aluminum and bismuth acted as surrogates in experiments to verify

that the plutonium samples would not "volatilize" and contaminate other equipment or become an inhalable health hazard. Diagnostic devices along the beamline and at the target chamber include thermal cameras and vacuum gauges. If the temperature inside rises too high or a vacuum failure is indicated, a valve at a bend in the beamline closes to stop the ion beam.

The team conducted several implantation experiments using the beamline. Samples were implanted with 1 atomic percent of helium (equivalent to the amount expected in 250-year-old plutonium). Irradiated material was then analyzed with a transmission electron microscope designed to accommodate actinides. With the Livermore microscope, researchers can examine minute samples and view features less than 1 nanometer across.

Micrographs of the samples showed the helium bubble size was similar to that observed in 40-year-old plutonium. Bubbles continued to grow when samples were annealed in an oven, but no damaging voids formed. “The oldest weapons in the stockpile are approximately 50 years old, but some scientists have wondered if there is a ‘cliff’ beyond which the pits would suffer deleterious effects,” says Bench. “Our research results may be an indication that the answer is no.” In fact, stockpile research completed several years ago determined that the credible lifetime for a plutonium nuclear weapon pit would be more than 85 years.

From NIF to the Newest Elements

In a Laboratory Directed Research and Development (LDRD) project, Bench, Brown, and Tumey are working with radiochemist Dawn Shaughnessy to use the ion beamline to produce radioactive gold tracers for analyzing debris collected from laser experiments at the National Ignition Facility (NIF). Debris diagnostics for NIF are conceptually similar to those previously used for forensics investigations following an underground nuclear experiment. The chemical and isotopic analysis techniques are the same.

At the center of the NIF target chamber, a tiny metal cylinder, or hohlraum, holds a spherical fuel capsule on which 192 laser beams are focused. When the fuel capsule explodes, a complex series of nuclear reactions takes place. Small devices inside the target chamber collect debris produced by these reactions. One role of the debris collectors is to record the number of nuclear activations. These data reveal how the capsule fuel mixes, the properties resulting from the fusion reaction, and any exotic nuclear decay processes. Understanding the complex reactions involved is relevant to issues concerning nuclear weapon performance.

“Debris comes from both the capsule and the hohlraum, but most of it is from the hohlraum,” says Shaughnessy. “Once

we’ve acquired good data on the hohlraum debris, our near-term plan is to embed various tracer isotopes in the fuel capsule, so we can differentiate the capsule and hohlraum data.”

In collaboration with the Colorado School of Mines and Los Alamos National Laboratory, experimenters are using NIF and smaller Livermore lasers to evaluate collector materials and determine the optimal distances for collector and blast shield placement. After an experiment, chemical processing involving the gold tracers will measure how much debris is generated with different materials and configurations. For example, tests with aluminum as a blast shield material showed it became badly cooked and melted, making it too dirty to use at a site in which cleanliness is a top priority.

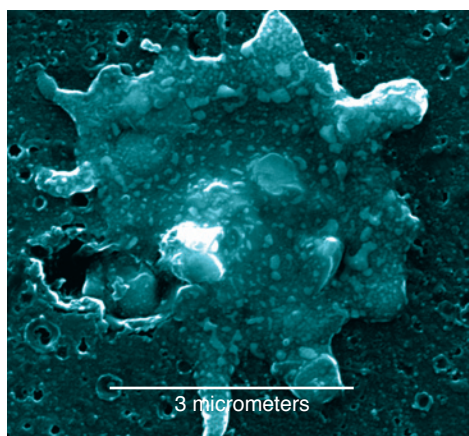
Debris analysis is currently performed in a radiochemistry laboratory. In the future, however, the highly sensitive measuring capability at CAMS may prove useful for this task.

In another LDRD-funded project, Shaughnessy and the CAMS team are planning to test rapid, automated chemistry systems connected to the ion beamline. The automated system will provide a tool for basic radiochemistry research. Potential applications for the future include providing real-time nuclear forensics capability in the field, producing medical isotopes, analyzing debris collected by NIF diagnostics, and characterizing the chemistry of any radioactive sample.

One application will examine the fundamental properties and chemical behavior of “superheavy” elements. The



Microscopist Mark Wall works with the Laboratory’s transmission electron microscope. With this microscope, researchers can examine tiny samples of irradiated material and view features less than 1 nanometer across.



The new high-energy ion beamline at CAMS produces radioactive gold tracers for measuring and analyzing debris collected in experiments at the National Ignition Facility (NIF). In one NIF experiment, a molten droplet of gold condensed quickly on the surface of an aluminum debris shield in the target chamber.

superheavy elements are those at the end of the periodic table and include six that were discovered by Livermore researchers in collaboration with colleagues at the Flerov Laboratory of Nuclear Reactions in Dubna, Russia. (See *S&TR*, October/November 2010, pp. 16–19; January/February 2002, pp. 16–23.)

Little is known about the recently discovered superheavy elements in part because so few atoms are produced before an element begins to decay. For example, researchers have yet to determine whether element 114, one of the Livermore–Dubna discoveries, is tetravalent or divalent, aqueous- or gas-phase. Because of the paucity of atoms, experiments to make these basic discoveries take a very long time. According to Shaughnessy, lighter elements with similar properties are used for chemistry development because they

have more atoms available for study. She notes that even with a few more atoms, an automated system is essential because experiments will require rapid chemistry with a high repetition rate performed over several weeks of beam time.

“Most accelerator managers don’t want to allocate beam time to time-consuming heavy-element work,” says Shaughnessy, “but Graham [Bench] was willing to support our project. And CAMS is the ideal location because it is onsite. We can transport samples between our radiochemistry laboratories and CAMS and modify the automated system as needed to optimize performance. In addition, team members with accelerator experience will perform the final system integration, which is scheduled for 2012.”

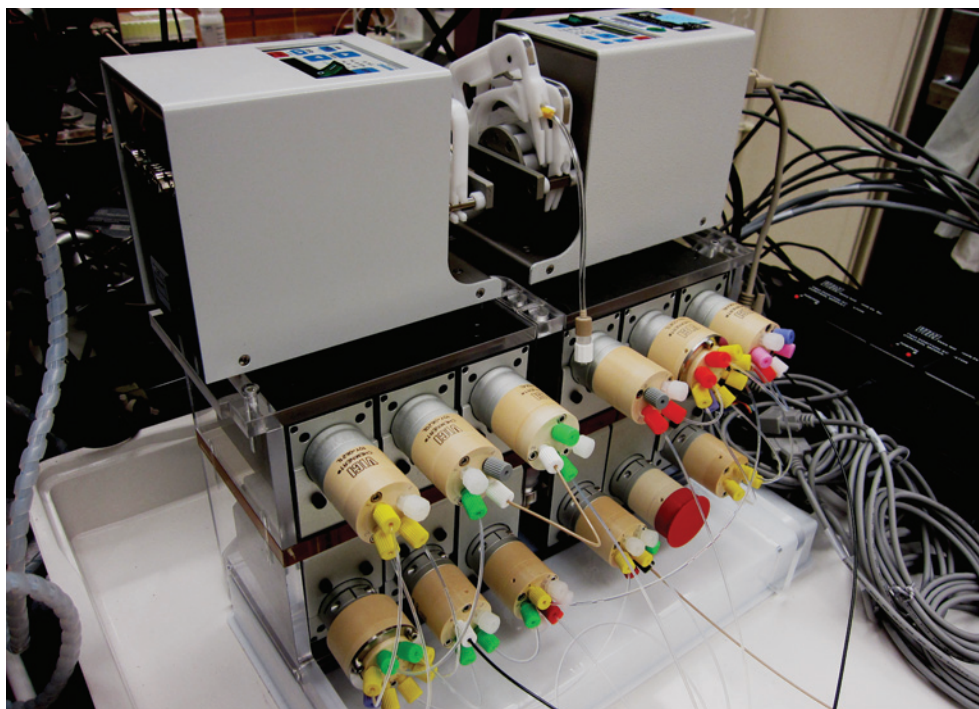
Developing the system's experimental setup and diagnostic processes is equally challenging because of the exceedingly small sample quantities. Shaughnessy and her colleagues are drawing on prior environmental work that involved tiny amounts of materials against a large background of other substances. They are investigating chemical separation and extraction techniques to remove interfering products and provide information about such properties as chemical bonding.

After final testing, the automated system will be shipped either to the Flerov Laboratory in Russia or to GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany, which operates a large-scale accelerator for heavy ions. Says Shaughnessy, "Collaborators at both sites have committed future beam time to extended heavy-element chemistry experiments."

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| Hydrogen 1 1.01 | | | | | | | | | | | | | | | | Helium 2 4.00 | | | | | | | | | | | | | | | |
| Lithium 3 6.94 | | Beryllium 4 9.01 | | | | | | | | | | | | | | Boron 5 10.81 | Carbon 6 12.01 | Nitrogen 7 14.01 | Oxygen 8 16.00 | Fluorine 9 18.99 | Neon 10 20.18 | | | | | | | | | | |
| Sodium 11 22.99 | | Magnesium 12 24.31 | | | | | | | | | | | | | | Aluminum 13 26.98 | Silicon 14 28.09 | Phosphorus 15 30.97 | Sulfur 16 32.06 | Chlorine 17 35.45 | Argon 18 39.95 | | | | | | | | | | |
| Potassium 19 39.10 | Calcium 20 40.08 | Scandium 21 44.96 | Titanium 22 47.87 | Vanadium 23 50.94 | Chromium 24 51.996 | Manganese 25 54.94 | Iron 26 55.85 | Cobalt 27 58.93 | Nickel 28 58.69 | Copper 29 63.55 | Zinc 30 65.38 | Gallium 31 69.72 | Germanium 32 72.63 | Arsenic 33 74.92 | Selenium 34 78.96 | Bromine 35 79.90 | Krypton 36 83.80 | | | | | | | | | | | | | | |
| Rubidium 37 85.47 | Sr 87.62 | Yttrium 39 88.91 | Zr 91.22 | Niobium 40 92.91 | Molybdenum 41 95.96 | Technetium 42 [98] | Ru 101.07 | Rh 102.91 | Pd 106.42 | Ag 107.87 | Cd 112.41 | In 114.82 | Sn 118.71 | Sb 121.76 | Te 127.60 | I 126.90 | Xenon 54 131.29 | | | | | | | | | | | | | | |
| Cesium 55 132.91 | Ba 137.33 | Lanthanum 57 138.91 | Hf 178.49 | Ta 180.91 | W 183.84 | Re 186.21 | Os 190.23 | Ir 192.22 | Pt 195.08 | Au 196.97 | Hg 200.59 | Tl 204.38 | Pb 207.20 | Bi 208.98 | Po [209] | At [210] | Rn [222] | | | | | | | | | | | | | | |
| Radium 87 [226] | Ra [226] | Ac~ [227] | Rf [261] | Db [268] | Sg [271] | Bh [272] | Hs [277] | Mt [278] | Ds [285] | Rg [286] | Cn [289] | Uu [294] | Uub [295] | Uut [296] | Uuh [297] | Uus [298] | Uuq [299] | | | | | | | | | | | | | | |
| Lanthanide Series | | | | | | | | | | | | | | | | | | Actinide Series | | | | | | | | | | | | | |
| Ce 58 140.12 | | | | | | | | | | | | | | | | | | Pr 59 140.91 | Nd 60 144.24 | Pm 61 [145] | Sm 62 150.36 | Eu 63 151.96 | Gd 64 157.25 | Tb 65 158.93 | Dy 66 162.50 | Ho 67 164.93 | Er 68 167.26 | Tm 69 168.93 | Yb 70 173.05 | Lu 71 174.96 | |
| Th 90 232.04 | | | | | | | | | | | | | | | | | | Pa 91 231.04 | U 92 238.03 | Np 93 [237] | Pu 94 [244] | Am 95 [243] | Cm 96 [247] | Bk 97 [247] | Cf 98 [251] | Es 99 [252] | Fm 100 [257] | Md 101 [258] | No 102 [259] | Lr 103 [262] | |

CAMS researchers are developing a rapid, automated chemistry system to examine the fundamental properties and chemical behavior of superheavy elements, including elements 113 through 118 (red) on the periodic table, which were discovered by a collaboration involving Lawrence Livermore and the Flerov Laboratory of Nuclear Reactions in Dubna, Russia.

Innovations at CAMS are also advancing research on the Laser Inertial Fusion Energy (LIFE) concept as a potential fusion power plant for the future. (See *S&TR*, July/August 2011, pp. 4–12.) NIF experiments are serving as a proving ground for LIFE, which



Livermore's automated chemistry system is designed for basic radiochemistry research. In the future, researchers may adapt the system to perform nuclear forensics in the field, produce medical isotopes, analyze debris collected by laser diagnostics, and characterize the chemistry of radioactive samples.

offers the promise of an unlimited supply of clean, sustainable energy. Materials inside a fusion reactor would be exposed to extremely energetic neutrons and temperatures of many hundreds of degrees. Selecting high-performance materials that can withstand this extreme environment is a challenge.

In conventional power plant construction, a system's design generally dictates the materials to be used. For a LIFE reactor, the availability of structural materials will affect the system design. Decisions regarding the allowable operating temperature, coolant, and power conversion system will depend on the performance characteristics of the construction materials.

Tumey and materials scientist Michael Fluss have developed methods for testing potential materials for a LIFE reactor by implanting them with various ions. Says Fluss, "We need radiation-tolerant materials, but they don't have to last for the 40- or 50-year life of the plant. They

would be constantly checked and replaced as needed, probably every 4 or 5 years."

According to Fluss, the optimal choice is a material that is already available and whose properties are well known. One such material is oxide-dispersion-strengthened (ODS) steel. This steel is produced commercially by mechanically alloying the elemental metallic powder with nanoparticles of yttria oxide and consolidating the material by hot extrusion or hot isostatic pressing. Thanks to the nanoparticles, ODS steels are relatively resistant to radiation-induced swelling. They are also stronger than conventional steels and more resistant to oxidation and corrosion at high temperatures.

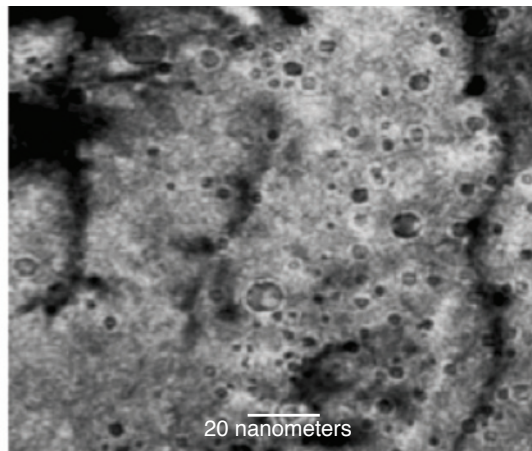
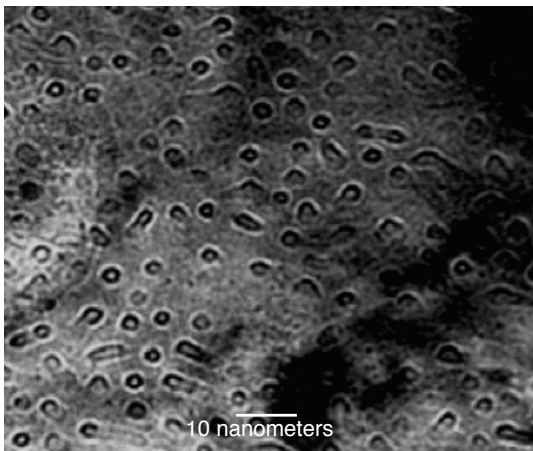
With no existing fusion power plants available for comparison purposes, other experimental platforms are the only route for testing LIFE materials. To mimic the conditions experienced by fusion materials, Fluss wants to simultaneously expose the iron-based steel to ion beams that implant iron, helium, and hydrogen.

These experiments are taking place in collaboration with researchers at CEA in Seclay, France, which provides three ion beams operating simultaneously from three electrostatic accelerators.

"The effects of helium are well known because of many years of experience with fission reactors," says Fluss. "Hydrogen is the wild card. Both gases are very light, but they behave differently. Helium is inert and moves slowly through steel's structural lattice. Hydrogen is chemically active and moves quickly through the lattice, interacting with helium and other defects to potentially create complex microstructures. Because a fusion reactor would have four times as much hydrogen as helium, it is imperative that we understand hydrogen's role and any deleterious effects associated with it."

In preparing for the experiments in France, Tumey and Fluss examined the role of the yttria nanoparticles. They investigated two steels, one containing nanoparticles and the other without, and exposed them to both iron and helium ion beams at CAMS. Livermore materials scientist Luke Hsiung then examined the samples using the Laboratory's transmission electron microscope.

High-resolution micrographs revealed that nanoparticles suppress radiation-induced swelling in the ODS steel. Helium-filled cavities tend to be trapped



To better understand how nanoparticles affect radiation-tolerant materials, Livermore researchers irradiated two steel samples with iron and helium ions. The sample with oxide-dispersed nanoparticles (left) formed small bubbles that remained similar in size. The sample without nanoparticles (right) shows a more varied distribution of cavities, some of which have started to grow into potentially damaging voids.

by nanoscale oxide particles and clusters. However, research by scientists in Japan has shown that when ODS steels are exposed to iron, helium, and hydrogen together, material swelling increases by a factor of 10. Says Fluss, “Experiments at CEA will help us better understand this problem with the goal of controlling it.”

Customer Community Growing

Livermore researchers led by materials scientist Wayne King are collaborating with Idaho and Argonne national laboratories on a proposal to study changes that occur in metallic fuels, a potential option for future fission power plants. To date, metallic fuels have been subject to less study than the more commonly used oxide fuels. Researchers have found, however, that fission gases released from the fuel could build up pressure and may crack the fuel’s

cladding. The tri-laboratory team plans to develop a predictive model for this process.

King, Tumey, and coworkers from Livermore and Argonne have found that developing the conditions to study helium bubbles in fuel is a challenge. In combination with an annealing process to mimic the appropriate temperatures to which fuel is subjected, xenon implantation needs very high energies of 80 megaelectronvolts.

Says King, “This project will be unique—if we receive funding, of course. It will combine not only theory, simulation, and experimentation but also uncertainty quantification, or UQ, which has only recently found its way into scientific predictions.” (See *S&TR*, July/August 2010, pp. 12–14.) King notes that measuring the degree of uncertainty is the only way to assess the quality of

model predictions. If this project proceeds, Tumey’s expertise at CAMS will again be put to work.

And the projects just keep coming. Says Bench, “With better tools and an expanding expertise in applying them to the challenges that come our way, CAMS is serving a growing community of customers. Our efforts on behalf of national security show no sign of slowing down.”

—Katie Walter

Key Words: accelerator mass spectrometry (AMS), Center for Accelerator Mass Spectrometry (CAMS), heavy-element chemistry, Laser Inertial Fusion Energy (LIFE), National Ignition Facility (NIF), national security, nonproliferation, nuclear forensics.

For further information contact Graham Bench (925) 423-5155 (bench1@llnl.gov).